

# Atmospheric lead deposition to Okefenokee Swamp, Georgia, USA

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**“Capsule”:** *Coal combustion emissions appear to be a major source of Pb in the Okefenokee wetland.*

## Abstract

Contamination of the environment from atmospheric deposition during the twentieth century is pervasive even in areas ostensibly considered pristine or remote from point sources. In this study, Pb concentrations in a <sup>210</sup>Pb-dated peat core collected from the Okefenokee Swamp, GA were used to assess historical contaminant input via atmospheric deposition. Lead isotope ratios were determined by dynamic reaction cell ICP-MS (DRC-ICP-MS). Increases in Pb concentration occurred in the late nineteenth century and a marked rise in Pb concentrations pre-dated the widespread use of leaded gasoline within the US. The <sup>206</sup>Pb/<sup>207</sup>Pb ratios of 1.19 during this period were consistent with coal combustion emissions. A later increase in Pb concentration, concurrent with a trend toward more radiogenic <sup>206</sup>Pb/<sup>207</sup>Pb ratios in gasoline is consistent with an increased input of Pb from leaded gasoline emissions. However, it appears that coal combustion emissions remain a major source of Pb to the Okefenokee.

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## 1. Introduction

The biogeochemical cycling of trace elements is significantly influenced by human activity, which for the most part has resulted in a redistribution of trace elements from the sub-surface (e.g. coal, oil, and mineral deposits) to near surface and surface environments. Trace element profiles in cores from peat bogs and lake sediments preserve a record of deposition that reflects both natural and anthropogenic inputs (Nriagu, 1979; Evans and Rigler, 1985; Graney et al., 1995; Farmer et al., 1997; MacKenzie et al., 1997; Shotyk et al., 1998, 2000b, 2002; Weiss et al., 1999; Nieminen et al., 2002; Weiss et al., 2002). The majority of studies have revealed

increases in trace element concentrations in the near surface that are consistent with anthropogenic inputs, although the relative enrichment of a particular trace element and importance of fluvial versus atmospheric deposition is site specific (Skei and Paus, 1979; Norton, 1986; Graney et al., 1995). Relatively pristine environments are not immune to the impact of anthropogenic emissions due to atmospheric deposition and Pb concentration profiles in ice cores from Greenland and Antarctica reflect the wide-ranging influence of industrial contamination (Boutron et al., 1994). Atmospheric deposition of Pb has been most intensively studied because this element has been used from antiquity, the primary pathway for its redistribution in the environment is atmospheric deposition (Nriagu and Pacyna, 1988) and the distinctive isotope ratios of naturally occurring Pb sources afford a unique opportunity to identify the actual source of Pb contamination (Sturges and Barrie, 1987). For example, Pb concentrations and <sup>206</sup>Pb/<sup>207</sup>Pb

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isotope ratios in a sectioned and dated core taken from a peat bog in the Swiss Jura Mountains revealed changes in Pb fluxes due to climatic changes 10,000 year BP and the prevalence of anthropogenic sources dating back 3000 year BP (Shotyk et al., 1998). The most pervasive source of Pb to the atmosphere has resulted from its use for most of the twentieth century as a gasoline additive. Many Western countries, however, banned leaded gasoline in the latter part of the twentieth century, and the subsequent decrease in atmospheric Pb deposition (Eisenreich et al., 1986), has been reflected in data from lake sediments and peat cores (Schell et al., 1992; Gobeil et al., 1995)

The Okefenokee National Wildlife Refuge is a Class I Air Quality Area, based on the 1977 Clean Air Act Amendments. The inflow of water to the swamp is predominantly via rainfall (82%), while surface runoff contributes 15% and groundwater inflow 3% (Patten and Matis, 1984). The swamp is a peatland and the nutrient supply is primarily from rainwater with minimal contribution from groundwater, hence it is considered to be ombrotrophic. The main vegetation of the Okefenokee Swamp include the aquatic plants: *Nuphar*, *Orontium*, *Eliocharis*, *Eriocaulon*, *Nymphaea* and *Utricularia* and the tree species *Taxodium*, *Cyrilla*, *Magnolia*, *Ilex*, and *Persea* (Casagrande and Erchull, 1976). Peat cores from ombrotrophic bogs are sensitive indicators of the history of anthropogenic deposition. It is reasonable to expect that the airshed for the Okefenokee may be negatively impacted by the proximity of a number of cities within 50 miles (Waycross, GA, Brunswick, GA, Jacksonville, FL) and their associated industries. In this study we report Pb concentration profiles and isotope ratios in a dated core taken from Bluff Lake in the Okefenokee swamp, GA.

## 2. Materials and methods

### 2.1. Site description

Okefenokee Nation Wildlife Refuge, established in 1937, is located in southeastern Georgia and encompasses 1600 km<sup>2</sup> of the Okefenokee Swamp (Eadie, 1984). The Okefenokee Swamp is the second largest swamp in the US and is located in southeastern Georgia and northern Florida (latitude of 82° 27' and longitude of 30° 55'). The swamp is 9–14 m above sea level and is developed on an interglacial Pleistocene marine sediment (Casagrande and Erchull, 1976). Two rivers drain the Okefenokee: the Suwannee and St. Mary's River. The watershed for the swamp is small, on the east and north east sides the drainage area averages only 1.6 km in width while about 1215 km<sup>2</sup> drain into the swamp on the north and northwest (Casagrande and Erchull, 1976). The soils of the Atlantic Coastal Plain are

predominantly sand, hence there is little erosion or siltation within the swamp. Bluff Lake is a permanently flooded area of Okefenokee refuge located in the northeast quadrant.

### 2.2. Sampling and analysis

A sediment core sample was collected from Bluff Lake on 28 October 1992. The core was collected using a thick-walled (7 mm) plexiglass tube that was 2 m long and 10 cm in diameter. Using a piston extruder calibrated at 1-cm intervals, the core was sectioned from the top (surface) using a stainless-steel spatula. The top 50 cm of the core was sectioned at 2 cm (from 0 to 4 cm) and 1 cm (from 4 to 50 cm) intervals. Seven sub-samples encompassing the top 25 cm of the core were submitted for <sup>210</sup>Pb dating using the constant rate of supply (CRS) model courtesy of Dr. Peter Appleby at University of Liverpool, UK. Percent moisture (drying at 105 °C) and % organic matter (ashing at 480 °C) were gravimetrically determined for the sectioned core samples. Percent moisture was relatively consistent throughout the core with an average of 93%, percent organic matter results are included in Table 2. For trace metal analysis approximately 200 mg (dry weight) of each core section was weighed and placed into a Teflon microwave digestion vessel and 9 ml of trace metal grade HNO<sub>3</sub> (Fisher Scientific, Fairlawn, NJ) and 1.5 ml HF were added. Digestion was carried out in a CEM (Mathews, NC) MARS 5 Microwave Digestion unit, which provides constant temperature and pressure sensing. The EPA method 3052 was used to define temperature and pressure limits and reaction times for the digestion procedure. One replicate of the standard reference material San Joachin soil (SRM 2709, NIST, Gaithersburg, MD) and four replicates (one per microwave digestion carousel) of the marine sediment certified reference material MESS 3 (CNRC, Ottawa, ON, Canada) were included for quality control. A blank and one sample duplicate were included in each digestion carousel. Digested samples were diluted to 50 ml with 18 MΩ-cm DI water and stored in 50-ml centrifuge tubes. The digested samples were further diluted five times with DI H<sub>2</sub>O prior to trace element analysis by inductively coupled plasma mass spectrometry (ICP-MS). Trace element analysis was conducted in accordance with EPA method 6020 in terms of the frequency and acceptance criteria of quality control parameters using the ICP-MS instrument Elan 6100 DRC (Perkin Elmer, Shelton, CT) operated in standard mode with a Rytan spray chamber and cross-flow nebulizer. Rhodium was used as an internal standard. The ICP-MS signal at 206, 207, and 208 was summed for Pb quantitation. All results are reported as mg kg<sup>-1</sup> dry weight. Percent recoveries for standard reference materials MESS-3 (*n* = 4) were: 99% (± 4%) and 98% (± 4%), for Ti, and Pb, respectively,

and for NIST SRM 2709 recoveries were 96 and 90%, for Ti and Pb, respectively. Average relative percent difference for the four duplicate sample digestions were: 6 and 10% for Ti and Pb, respectively. Enrichment factors (EF) for Pb were calculated by taking the Pb/Ti concentration ratio of each core section and referencing that ratio to the upper crustal abundance ratio for these elements (Wedepohl, 1995). Lithogenic contributions to the total Pb burden of the sediment were calculated as the product of the Ti concentration and the Pb/Ti upper crustal abundance. Anthropogenic Pb was then calculated from the difference of total Pb and lithogenic Pb (Shoty et al., 2002).

Isotope ratios for Pb were determined on the acid digests by dynamic reaction cell ICP-MS (Elan 6100 DRC-ICP-MS) using the instrumental parameters in Table 1. Collisional focusing and thermalization of ions in the reaction cell can increase the precision of isotope ratio determination when compared to standard mode quadrupole ICP-MS (Bandura and Tanner, 1999; Bandura et al., 2000). Mass bias in the ICP-MS was corrected by running NBS 981 certified reference material. Results for NBS 981 were within 1% of the certified values. Average precision for the isotope ratio analysis reported herein was <0.1% relative standard deviation (RSD) when  $^{206}\text{Pb}$  and  $^{207}\text{Pb}$  signal intensity were 200,000 counts per second, which equates to approximately  $25 \mu\text{g l}^{-1}$  Pb in the analysis solution. This is a considerable improvement over conventional quadrupole ICP-MS isotope ratio precision. However, lower count rates, as a result of lower Pb concentration in the digestate, gave lower precision isotope ratio measurements of <0.5% RSD for count rates of 10,000 counts per second ( $1\text{--}2 \mu\text{g l}^{-1}$  Pb).

### 3. Results and discussion

Trace element concentration profiles in peat cores and lake sediments as a function of depth have been used to make inferences concerning contaminant inputs.

Table 1  
Dynamic reaction cell (DRC)-ICP-MS operating parameters for Pb isotope ratio measurements

DRC-ICP-MS operation conditions	
Plasma gas flow	15 l/min
Auxiliary gas flow	1.2 l/min
Nebulizer gas flow	0.85 l/min
RF power	1100 W
DRC cell gas	$\text{H}_2/\text{Ar}$
DRC cell gas flow rate	0.5 ml/min
Sweeps/reading	500
Readings/replicate	20
Replicates	10
Dwell time per amu	1 msec
Isotopes measured	$^{206}\text{Pb}$ , $^{207}\text{Pb}$ , $^{208}\text{Pb}$

However, the sediment record can be confounded by variations in sedimentation rates, sediment focusing and post depositional remobilization processes such as resuspension and bioturbation. Nevertheless, there is now a substantial body of evidence that Pb, at least, is effectively immobile in peat profiles and that ombrotrophic bogs preserve a record of the history of anthropogenic inputs of Pb (Shoty et al., 2000a). Lead concentrations, Ti concentrations and the calculated Pb contributions from lithogenic and anthropogenic origin are listed in Table 2.

Lead concentrations exhibited the most dramatic increase in concentration in the upper sections of the core (Fig. 1c). Ambient, pre-industrial concentrations of Pb in the sediments were about 0.5 mg/kg. Concentrations began to rise markedly around 1870 reaching a maximum concentration of about 31 mg/kg at a  $^{210}\text{Pb}$  age ca. 1935 and remained at this concentration until after 1965. This maximum represents an increase over background Pb concentrations of approximately 80 times and an enrichment factor of 18. Lead concentrations in the most recent sections of the core decreased from this maximum, presumably reflecting an increase in air quality due to the enactment of the Clean Air Act in the US in 1970. The primary anthropogenic source of Pb to the environment is through atmospheric deposition and the decrease in recent Pb inputs reflected in the sediment core are in accordance with the low concentrations of Pb ( $<3.3 \mu\text{g l}^{-1}$ ) recorded in rainfall at the Okefenokee between 1993 and 1995 (Winger and Lasier, 1997).

Consistent with the higher ash content of the upper sections of the core, Ti concentrations also increased, hence the calculated lithogenic Pb (Fig. 1a) contribution to total Pb (Fig. 1c) in the core increased in the upper (more recent) sections, presumably representing an increase in either aeolian or fluvial particulate input. Notwithstanding this increase in lithogenic Pb, the major contribution to the increase Pb was through anthropogenic Pb (Fig. 1b) inputs to the Okefenokee.

Similar concentration profiles for Pb have been reported for other lake sediments and peat bogs in the North America as well as worldwide (Nriagu, 1979; Graney et al., 1995; Weiss et al., 2002). In a study of 10 Adirondack lakes, Pb maximum concentrations ranged from 270 to 759 mg kg $^{-1}$ , the year of maximum accumulation ranged from 1967 to 1982 and enrichment factors ranged from 15 to 67 (Norton, 1986). Similarly, Pb in Great Lakes sediments had maximum concentrations 108, 124, and 61 mg/kg for Lakes Michigan, Ontario, and Erie, respectively, with peak maximums occurring during 1960–1977 (Graney et al., 1995). The much higher background and maximum Pb concentrations in sediments from the Great Lakes and Adirondack Lakes compared with the results reported herein for the Okefenokee reflects both a greater influence of

Table 2  
 $^{210}\text{Pb}$  dates, % organic matter, Pb and Ti concentrations,  $^{206}\text{Pb}/^{207}\text{Pb}$  isotope ratio, Pb enrichment factor and relative lithogenic and anthropogenic Pb contribution to total Pb for individual core sections

Depth (cm)	$^{210}\text{Pb}$ date	Organic matter (%)	Pb total (mg/kg)	Ti (mg/kg)	Pb/Ti	$^{206}\text{Pb}/^{207}\text{Pb}$	Pb EF	Pb (mg/kg) lithogenic	Pb (mg/kg) anthropogenic
0–2	1990	77.56	18.08	257.8	0.0701	1.200	12.86	1.41	16.68
2–4		78.14	20.60	251.0	0.0821	1.199	15.05	1.37	19.23
5–6	1980	78.89	23.73	256.5	0.0925	1.204	16.96	1.40	22.33
6–7		91.17	26.85	285.1	0.0942	1.206	17.27	1.56	25.30
7–8		80.03	28.99	290.5	0.0998	1.202	18.30	1.58	27.41
8–9	1965	79.95	31.17	315.9	0.0987	1.203	18.09	1.72	29.44
9–10		80.15	30.09	291.3	0.1033	1.201	18.94	1.59	28.50
10–11		81.37	29.62	289.4	0.1024	1.197	18.77	1.58	28.04
11–12		82.26	31.73	295.0	0.1076	1.194	19.72	1.61	30.12
12–13	1935	83.56	24.66	278.1	0.0887	1.192	16.26	1.52	23.14
13–14		87.45	17.66	265.1	0.0666	1.191	12.22	1.45	16.22
14–15		85.25	15.20	283.0	0.0537	1.191	9.85	1.54	13.66
15–16		87.23	10.05	287.8	0.0349	1.191	6.40	1.57	8.48
16–17	1905	88.75	6.93	177.1	0.0391	1.191	7.17	0.97	5.96
17–18		88.61	5.73	192.5	0.0298	1.197	5.46	1.05	4.68
18–19		91.37	3.19	112.4	0.0284	1.199	5.20	0.61	2.58
19–20		91.39	2.25	97.6	0.0230	1.214	4.22	0.53	1.72
21–22	1870	94.16	1.11	106.0	0.0105	1.198	1.93	0.58	0.54
23–24		94.07	0.63	106.7	0.0059	1.212	1.09	0.58	0.00
27–28		94.77	0.50	92.2	0.0054	1.249	0.99	0.50	0.00
28–29		94.83	0.51	170.8	0.0030	1.220	0.54	0.93	0.00
29–30		94.48	0.53	150.9	0.0035	1.213	0.64	0.82	0.00
31–32		94.57	0.53	118.8	0.0045	1.210	0.82	0.65	0.00
32–33		94.65	0.55	126.4	0.0044	1.212	0.80	0.69	0.00
34–35		93.69	0.60	149.7	0.0040	1.209	0.73	0.82	0.00
36–37		93.84	0.58	144.6	0.0040	1.208	0.73	0.79	0.00
37–38		93.59	0.49	124.9	0.0039	1.213	0.72	0.68	0.00
38–39		93.08	0.56	152.4	0.0037	1.211	0.68	0.83	0.00

erosional input to the sediments and greater atmospheric emissions from the more populated and industrial northeastern US. The Pb concentration profile in a core taken from a peat bog in eastern Canada exhibited a similar low background concentration ( $0.9 \pm 0.5 \text{ mg kg}^{-1}$ ) as that for Okefenokee background Pb values (Weiss et al., 2002), but the maximum concentration of  $52.5 \text{ mg kg}^{-1}$  is much greater than that reported here for the Okefenokee. However, the maximum EF, compared to either the average upper crustal abundance ratio for Pb/Ti or to the respective pre-industrial Pb/Ti ratio is greater for the Okefenokee, where the increase in EF is approximately  $25\times$  greater than pre-industrial values, but only  $10\times$  for the eastern Canada peat bog. This suggests a greater anthropogenic Pb input to the Okefenokee compared with the eastern Canada site.

### 3.1. Lead isotope ratios

The relative composition of the stable isotopes of Pb presents an effective method to identify the source of anthropogenic inputs of Pb to a system because Pb isotope ratios differ significantly dependent on the source. Isotope ratios for  $^{206}\text{Pb}/^{207}\text{Pb}$  determined in the sediment core from Okefenokee are shown in Fig. 2. The overall variation in the  $^{206}\text{Pb}/^{207}\text{Pb}$  throughout the

core was not large, ranging from 1.19 to 1.21, and this range is approaching the limit of precision for quadrupole DRC-ICP-MS isotope ratios to show significant difference between samples. However a significant trend over time was apparent using the quadrupole DRC-ICP-MS values. Pre-industrial  $^{206}\text{Pb}/^{207}\text{Pb}$  ranged from 1.2 to 1.21, but this ratio began to decrease to less radiogenic values around the first  $^{210}\text{Pb}$  dated section of the core (1870). The lowest  $^{206}\text{Pb}/^{207}\text{Pb}$  values of ca. 1.195 were obtained during 1900–1965 and after this time  $^{206}\text{Pb}/^{207}\text{Pb}$  began to rise again to values of 1.2 by 1990. The relatively rapid decrease in  $^{206}\text{Pb}/^{207}\text{Pb}$  from 1870 to 1905 is consistent with a marked increase in anthropogenic Pb to the Okefenokee during this period, which, due to the low pre-industrial Pb values, rapidly became the major input source of Pb to the Okefenokee.

For most of the nineteenth century, the Okefenokee and surrounding area were sparsely populated; however, beginning in the late nineteenth century with the Suwannee Canal Company and continuing into the twentieth century with the Hebard Lumber Company, significant commercialization of the swamp, predominantly for timber production, began. The logging companies established a network of railroads to facilitate the transport of lumber out of the interior of the swamp. Comparing this historical perspective with that of the sediment Pb

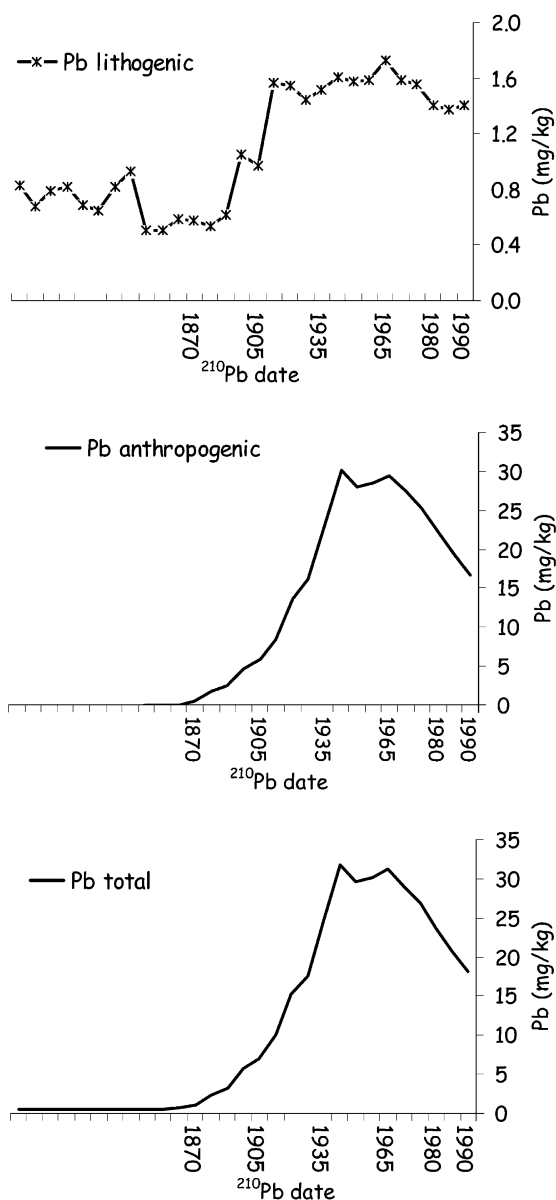


Fig. 1. Concentration profile for (a) lithogenic Pb, (b) anthropogenic Pb and (c) total Pb.

record (Fig. 1) and the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios (Fig. 2) is instructive. Sediment Pb concentrations exhibited a rapid increase from 1870 to 1935 (Fig. 1), concurrently, Pb isotope ratios fell from  $>1.2$  before ca. 1870 to 1.193 at the beginning of the twentieth century. Given the importance of the railroad in these logging operations, it follows that coal burning would have contributed significant local contamination to the Okefenokee during this time period. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios for West Virginia and Pennsylvania coal, the major US sources at this time, are 1.212–1.18 (Chow and Earl, 1972). Thus increased input of Pb to the Okefenokee from coal burning is consistent with the observed decrease in  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios observed in the core for this time period.

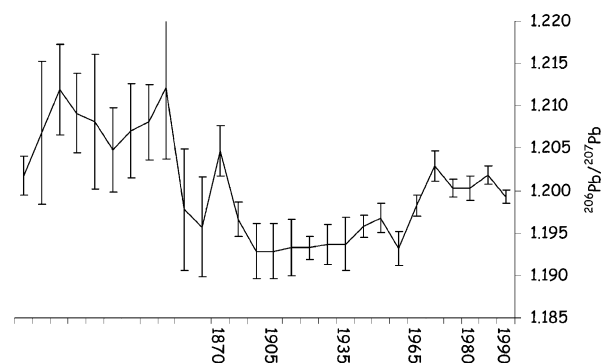


Fig. 2.  $^{206}\text{Pb}/^{207}\text{Pb}$  isotope ratio profile in  $^{210}\text{Pb}$ -dated Okefenokee sediment core. Error bars are one standard deviation.

In 1937, the Okefenokee was designated a National Wildlife Refuge and commercial logging activities essentially ceased. On a national scale, after 1930, the major Pb emission to the atmosphere was from leaded gasoline (Graney et al., 1995). The Pb in the peat core record from the Okefenokee exhibits a further decrease around 1965 that is consistent with a different input source of Pb to the swamp, i.e. leaded gasoline emissions. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios during this time remain relatively constant; however, they shift to more radiogenic values in the 1960s concomitant with a fall in total Pb concentrations from this time to the present. The isotopic signature of leaded gasoline changed throughout the twentieth century. Prior to 1967 the mean  $^{206}\text{Pb}/^{207}\text{Pb}$  value was estimated to be 1.153 (Erel and Patterson, 1994), but this ratio increased after 1968 with the increasing use of the more radiogenic Missouri Pb ore ( $^{206}\text{Pb}/^{207}\text{Pb} > 1.35$ ) producing  $^{206}\text{Pb}/^{207}\text{Pb}$  values of around 1.23. The fact that the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio at Okefenokee remained relatively constant during 1930–1960 at 1.195, despite the predominant gasoline  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio being significantly lower than this suggests that local Pb emissions continued to be the major source of Pb to the Okefenokee in this time period. However, the small rise in the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio in the core after 1965 is consistent with the input of Pb derived from gasoline emissions from the more radiogenic Missouri ore.

The phasing out of leaded gasoline in the US in the 1970s coupled with the designation of the Okefenokee as a class 1 airshed has significantly decreased Pb deposition to the Okefenokee (Fig. 1). The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.2 found in peat from the Okefenokee Swamp dated in the 1980s is consistent with the typical value reported for US aerosols during that time i.e.  $\sim 1.19$  to 1.21 (Church et al., 1990).

#### 4. Conclusions

Elevated concentrations of Pb were identified in peat core sections taken from the Okefenokee National

Wildlife Refuge that corresponded to increased atmospheric deposition of Pb during the twentieth century. Isotope ratio analysis of  $^{206}\text{Pb}/^{207}\text{Pb}$  in the core sections suggests that coal combustion was the primary source of Pb to the Okefenokee. The general enactment of the Clean Air act within the US, the specific designation of the Okefenokee as a Class 1 Air Quality Area, and restrictions of Pb in gasoline appear to have been effective in reducing the contaminant loading of Pb to the Okefenokee Swamp through atmospheric deposition.

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